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(71)Applicant : **ASAHI CHEM IND CO LTD**

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(72)Inventor : **MIYAMOTO AKIRA  
SATO MASAKAZU**

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### (54) PROPYLENE RESIN MOLDING WITH IMPROVED COATABILITY AND PRODUCTION THEREOF

(57)Abstract:

**PURPOSE:** To improve the coating suitability without impairing the balance among moldability, cost, mechanical properties, chemical resistance, electrical properties, etc., of a propylene resin molding by thermally plasticizing a specific resin compsn. and cooling it under pressure.

**CONSTITUTION:** 100 pts.wt. resin component comprising 99.9-10 pts.wt. hydroxylated polypropylene resin having a hydroxyl content of 0.1×1-2-50 meq/g and 0.1-90 pts.wt. olefin elastomer is compounded with 0.01-10 pts.wt. material comprising organotin compd. and/or a tert. amine compd. The resultant resin compsn. is charged into a mold cavity having the surface coated with a material having a thermal conductivity of 0.01cal/c.sec.C (at 20°C) or lower and a surface tension of 35mN/m (at 20°C) or higher, thermally plasticized, extruded, and cooled under pressure.

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DETAILED DESCRIPTION

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[Detailed Description of the Invention]

[0001]

[Industrial Application] This invention relates to the propylene resin Plastic solid excellent in the paintwork-ed over an urethane system coating. The propylene resin Plastic solid of this invention is the field of autoparts, electric product components, a machine part, a toy, stationery, daily needs, etc., and is preferably used for the application for which especially paint and adhesion are needed.

[0002]

[Description of the Prior Art] Since property balance, such as a moldability, cost, mechanical characteristics and chemical resistance, and an electrical property, is excellent, propylene resin is used in the field of autoparts, the electrical and electric equipment and electronic parts, a machine part, etc. However, since propylene resin does not have a polar group in structure, it has the difficulty in paint film adhesion, an adhesive property, etc., and it has been a failure on the real application exploitation with these new.

[0003] Wet surface treatment methods, such as the approach of processing using mineral acids, such as dry type surface treatment methods, such as a flame treatment method, a plasma treatment method, an ozone approach, a corona discharge approach, ultraviolet rays, or an electron-beam-irradiation approach, chromic acid mixture, and concentrated sulfuric acid, as a means for improving the paint film adhesion of propylene resin and an adhesive property and an approach to which a Plastic solid front face is made to carry out the graft of the surface treatment component chemically, or the primer coating method which applies a surface treatment layer directly is mentioned. However, these approaches were not desirable approaches from the problem on a facility, or productivity in the operation.

[0004] By using a peroxide for propylene resin and on the other hand, carrying out graft denaturation of the polar-group content compound, there is a method of improving the paintwork-ed of propylene resin, and since it says that these approaches have a processing facility and simple down stream processing, economic merit is high and attracts attention especially in recent years. However, since the chain cutting reaction of propylene resin advanced with a graft reaction, mechanical physical properties, such as rigidity, shock resistance, and thermal resistance, fell, for this reason the activity might be restricted. Moreover, in order to acquire sufficient paint film adhesion force, when the amount of grafts of a polar-group content compound is increased, the problem which the increment in a bleeding object, generating of an odor, change of a color tone, etc. do not have may arise.

[0005]

[Problem(s) to be Solved by the Invention] This invention is made in order to acquire the propylene resin Plastic solid which has the paintwork-ed which was excellent, without spoiling property balance which propylene resin has, such as a moldability, cost, mechanical characteristics and chemical resistance, and an electrical property.

[0006]

[Means for Solving the Problem] this invention persons found out that there was a correlation in the contact angle and paint film adhesion of the waterdrop to a Plastic solid front face, as a result of

examining acquiring the propylene resin Plastic solid excellent in the paint film adhesion force and adhesive strength. That is, it was the resin constituent Plastic solid which consists of a hydroxyl-group content polypropylene resin which contains a hydroxyl group by the specific density range in a polypropylene molecule, and when the contact angle of the waterdrop to this Plastic solid front face was 80 degrees or less, it found out that paintwork-ed improved remarkably. When fabricating using the mold which has a further specific front face, it found out that refining which was excellent in paint film adhesion, without being able to advance efficiently orientation by the side of the Plastic solid front face of a polar group by making the interface affinity of a polar-group-die front face into driving force, for this reason hardly spoiling productivity in the simple nature of the fabricating operation of propylene resin and a list could be obtained. Furthermore, since the amount of grafts of a polar-group content compound is excellent in paintwork-ed at least, the Plastic solid of this invention can solve problems, such as lowering of the various physical properties accompanying chemistry denaturation, and an increment in a bleeding object, generating of an odor, change of a color tone.

[0007] That is, the propylene resin Plastic solid excellent in the paintwork-ed and the adhesive property of this invention is a Plastic solid of the resin constituent which consists of the following (a), (b), and (c) components, and is a propylene resin Plastic solid characterized by the contact angle of the waterdrop to this Plastic solid front face being 80 degrees or less.

A hydroxyl group Component : (a) More than  $0.1 \times 10^{-2}$  meq/g (PP) The hydroxyl-group content polypropylene resin 99.9 contained below 50 meq/g (PP) - 10 weight section (b) component: Olefin system elastomer ( ) 90 [ 0.1 - ] weight section (c) component: An organotin compound and/or tertiary amine compound a) -- a total of 100 weight sections of a component and the (b) component -- receiving -- 0.01 - 10 weight section -- as an approach of acquiring the Plastic solid which has such a front face The approach of acquiring a Plastic solid by supplying the ingredient whose surface tension is more than 35 mN/m (20 degrees C) about the resin constituent which consists of the above-mentioned component to the mold for resin shaping which it has on a mold cavity front face, and carrying out a melting press, Or after heating beforehand the resin molding die which has the ingredient whose surface tension is more than 35 mN/m (20 degrees C) on a mold cavity front face to an elevated temperature 100 degrees C or more, the resin by which heating plasticization was carried out can be supplied, and the shaping approach which carries out application-of-pressure cooling can be mentioned.

[0008] Furthermore, the thermal conductivity of the Plastic solid which has this Plastic solid front face is below 0.01 cal/cm-sec and \*\* (20 degrees C). The ingredient whose surface tension is more than 35 mN/m (20 degrees C) and in the mold cavity for resin shaping which it has on a mold cavity front face The resin constituent which consists of the above-mentioned component by which heating plasticization was carried out can be supplied, and it can obtain also by the approach of carrying out application-of-pressure cooling, and according to this approach, the front face which was excellent in paintwork-ed, without hardly spoiling productivity in the simple nature of the fabricating operation of resin and a list can be obtained.

[0009] Hereafter, this invention is explained to a detail.

(1) The Plastic solid of resin constituent this invention consists of resin constituents which consist of the following (a), (b), and (c) components.

A hydroxyl group Component : (a) More than  $0.1 \times 10^{-2}$  meq/g (PP) The hydroxyl-group content polypropylene resin 99.9 contained below 50 meq/g (PP) - 10 weight section (b) component: Olefin system elastomer ( ) 90 [ 0.1 - ] weight section (c) component: An organotin compound and/or tertiary amine compound a) (a) for the propylene resin Plastic solid of 0.01 - 10 weight section this invention to discover the firm paint film adhesion force to a total of 100 weight sections of a component and the (b) component and the rate of (b) are 1 - 90 weight sections to 99 - 10 weight section preferably.

Furthermore, it is good to choose out of the range of 10 - 70 weight section to 90 - 30 weight section preferably. moreover, the loadings with desirable tin compound and/or tertiary amine compound (c) component -- a total of 100 weight sections of the (a) component and the (b) component -- receiving -- 0.05 - 5 weight section -- it is 0.1 - 3 weight section still more preferably.

[0010] The component which constitutes hereafter the resin constituent used by this invention is

explained to a detail.

(a) With the hydroxyl-group content polypropylene resin which constitutes the resin constituent used by hydroxyl-group content polypropylene resin this invention The denaturation polypropylene which carried out the graft of at least one sort of unsaturated carboxylic acid, or the partial saturation dicarboxylic acid anhydride The organic compound with which it receives (it is hereafter described as acid denaturation polypropylene), and one or more pieces and one or more hydroxyl groups are contained for the first-class amino group in a monad It can obtain by making it react (it is hereafter written as a hydroxy group content amine compound). More than  $0.1 \times 10^{-2}$  meq/g (PP), a hydroxyl-group content is below 50 meq/g (PP), and if the amount of hydroxyl groups is in this range, it can also blend and use non-denaturalized polypropylene for the denaturation polypropylene obtained by the reaction. In addition, the hydroxyl-group content shown here is a content of the hydroxyl group introduced into the polypropylene molecule by the chemical bond, and the hydroxyl group contained in the unreacted hydroxy group content amine compound which remains in a resin constituent is excepted. Since the paintwork-ed amelioration effectiveness is not acquired for a hydroxyl-group content below by  $0.1 \times 10^{-2}$  meq/g (PP), but the mechanical strength of a Plastic solid is spoiled above 50 meq/g (PP) or many bleeding objects are generated on a Plastic solid front face on the other hand, it is not desirable. The desirable hydroxyl-group contents of a hydroxyl-group content polypropylene resin are  $0.5 \times 10^{-2}$  - 10 meq/g (PP), and are  $1 \times 10^{-2}$  - 5 meq/g (PP) still more preferably.

[0011] The manufacture approach of a hydroxyl-group content polypropylene resin of having hereafter the hydroxyl group which uses acid denaturation polypropylene as a raw material is explained in detail.

(a-1) The acid denaturation polypropylene used for manufacture of an acid denaturation polypropylene hydroxyl-group content polypropylene resin is obtained by carrying out the graft of unsaturated carboxylic acid or the partial saturation dicarboxylic acid anhydride to polypropylene by the well-known approach.

[0012] The constituent concentration of the unsaturated-carboxylic-acid radical contained in acid denaturation polypropylene or a partial saturation dicarboxylic acid anhydride radical is 0.01 - 10 % of the weight, and especially its 0.1 - 5 % of the weight is desirable. Adhesive amelioration cannot be attained in the paintwork-ed list which is the object of this invention as the constituent concentration of an unsaturated-carboxylic-acid radical or a partial saturation dicarboxylic acid anhydride radical is 0.01 or less % of the weight. Since the mechanical strength of the resin constituent eventually obtained with it being 10 % of the weight or more on the other hand is spoiled greatly, it is not desirable.

[0013] The polypropylene used for manufacture of acid denaturation polypropylene is the homopolymer of a propylene, or the copolymer of a propylene and other alpha olefins. A propylene and the alpha olefin to copolymerize For example, ethylene, Butene-1, a pentene 1, the 2-methylbutene 1, the 3-methylbutene 1, a hexene 1, 3-methyl pentene 1, 4-methyl pentenes 1 and 3, 3-dimethyl butene-1, a heptene 1, methylhexane 1, the dimethyl pentene 1, trimethyl butene-1, The ethyl pentene 1, octene 1, the methyl pentene 1, the dimethyl hexene 1, the trimethyl pentene 1, the ethyl hexene 1, the methylethyl pentene 1, diethyl butene-1, the propyl pentene 1, DESEN 1, methyl NONEN 1, dimethyl octene, The trimethyl heptene 1, the ethyl octene 1, the methylethyl heptene 1, the diethyl hexene 1, dodecen 1, hexa dodecen, etc. can be mentioned. A random copolymer is sufficient as the copolymer of these alpha olefins and propylenes, and a propylene-alpha olefin block copolymer is sufficient as it. These alpha olefins can also use together one sort or two kinds or more. Moreover, as for the content of the alpha olefin in polypropylene, it is desirable that it is less than [ 45 mol % ]. As polypropylene used by this invention, a propylene homopolymer, the crystalline propylene ethylene block copolymer whose ethylene content is 2-40-mol %, and the crystalline ethylene propylene random copolymer whose ethylene content is 0.5-10-mol % are [ among these ] desirable. the melt index which measured such polypropylene by 230 degrees C and 2.16kg pile -- 0.05-100g/-- the thing of the range for 0.1-40g / 10 minutes is excellent in a moldability, and especially suitable for 10 minutes. Moreover, one kind of these polypropylene may be used and it may be used combining two or more kinds. Although it can manufacture by various approaches, such polypropylene can be made to be able to react to the bottom of existence of the combination catalyst of a solid-state-like titanium catalyst component and an organic

metal catalyst component, and can be obtained, for example.

[0014] Moreover, as the unsaturated carboxylic acid which carries out a graft to acid denaturation polypropylene, or a partial saturation dicarboxylic acid anhydride, dicarboxylic acid, such as carboxyl group \*\*\*\*\*, such as an acrylic acid, a methacrylic acid, a crotonic acid, a cinnamic acid, and an itaconic acid, a maleic acid, a fumaric acid, an itaconic acid, a chloro maleic acid, a citraconic acid, allyl compound succinic acid, mesaconic acid, and aconitic acid, and these acid-anhydride \*\*s are mentioned, for example. A maleic anhydride, an acrylic acid, and a methacrylic acid are desirable in these, and a maleic anhydride is the most desirable also in especially inside. These monomers can use together independent one sort or two sorts or more. Moreover, the amount of addition of unsaturated carboxylic acid or a partial saturation dicarboxylic acid anhydride can be increased by making a partial saturation aromatic series monomer live together, and making it react at a specific rate as indicated by JP,1-236214,A.

[0015] The approach of well-known arbitration can be used for manufacturing acid denaturation polypropylene. That is, the approach (solution method) of making an organic solvent carrying out the heating dissolution of the polypropylene, and making unsaturated carboxylic acid or a partial saturation dicarboxylic acid anhydride reacting to the bottom of existence of a radical generating agent, the method (scorification) of carrying out heating temperature up to the temperature more than the melting point, carrying out melting to it, and making unsaturated carboxylic acid or a partial saturation dicarboxylic acid anhydride react to existence of a radical generating agent, and the radiation graft method for using an electron ray etc. are employable.

[0016] In a solution method, it is desirable as an organic solvent to use aromatic solvents, such as a xylene, reaction temperature is performed at 100-180 degrees C, this approach has little side reaction and there is the description which can obtain the denaturation object with which unsaturated carboxylic acid or a partial saturation dicarboxylic acid anhydride was added to homogeneity. On the other hand, in the case of scorification, the extruder of a Banbury mixer, a kneader, one shaft, or a multiple spindle etc. can be used, and it can be made to react at the temperature of 300 degrees C or less more than the melting point of raw material resin. Scorification can terminate a reaction for a short time the top where actuation is easy. Although it is desirable to mix each of each resinous principles to homogeneity with a tumbler or equipment like a Henschel mixer beforehand in the state of powder or a pellet on the occasion of kneading, when required, mixing can be excluded and the approach of carrying out constant feeding to kneading equipment separately, respectively can also be used.

[0017] Especially although the radical generating agent used for a reaction can be suitably chosen from well-known things, its organic peroxide is desirable. As a desirable example, benzoyl peroxide, dicumyl peroxide, Lauroyl peroxide, 2, the 5-dimethyl -2, 5-G (tert-butyl peroxide) hexane, 2, the 5-dimethyl -2, 5-G (tert-butyl peroxide) hexyne - 3, 1, 3-bis(t-butyl PAOKI seesaw propyl) benzene, 1 and 1-screw (tert-butyl peroxide) - 3, 3, 5-trimethylcyclohexane, n-butyl -4, 4'-bis(tert-butyl peroxide) valerate, P-chlorobenzoyl peroxide, 2,4-dichlorobenzyl peroxide, T-butylperoxyisopropylcarbonate, di-t-butyl peroxide, t-butyl peroxybenzoate, a cumene hydroperoxide, diacetyl peroxide, lauroyl peroxide, t-butyl cumyl peroxide, etc. are mentioned.

[0018] Moreover, in order to remove by-products, such as unreacted components (unsaturated carboxylic acid or a partial saturation dicarboxylic acid anhydride, radical generating agent, etc.), oligomer of those, and a decomposition product, from a resultant, the approach of making it deposit and generating by the vent line, near an outlet in the middle of an extruder, after drawing in with a vacuum pump or dissolving a resultant in a suitable solvent can be used.

[0019] By-products, such as an unreacted component, oligomer, and a decomposition product, are also removable by heat-treating the acid denaturation propylene furthermore obtained at the temperature of 60 degrees C or more, or carrying out vacuum suction under melting.

(a-2) The approach of well-known arbitration can be used for manufacturing a hydroxyl-group content polypropylene resin from the acid denaturation polypropylene of the manufacture approach above of a hydroxyl-group content polypropylene resin. That is, the approach (solution method) of making an organic solvent carrying out the heating dissolution, and making acid denaturation polypropylene and a

hydroxy group content amine compound reacting to it, and the approach (scorification) of carrying out heating temperature up to the temperature more than the melting point, making carry out melting to it, and making it react are employable.

[0020] As an example desirable as a hydroxy group content amine compound used when manufacturing a hydroxyl-group content polypropylene resin 2-aminoethanol, 3-friend no 1-propanol, a 4-friend no 1-butanol, A 5-friend no 1-pentanol, a 6-friend no 1-hexanol, a 2-friend no 1-butanol, 2-(2-aminoethoxy) ethanol, N-aminoethylethanolamine, A 2-friend no 3-MECHIRU 1-butanol, 2-friend no 2-MECHIRU 1-propanol, 2-friend no 1-propanol, 3-friend no 1, 2-propanediol, 3-friend no 1,3-propanediol, 2-friend no 2-MECHIRU 1,3-propanediol, 2-(3-aminopropyl amino) ethanol, 2-friend no 2-ECHIRU 1,3-propanediol, Tris (hydroxyl methyl) aminomethane, o-, p-, m-aminophenol, 2-amino phenethyl alcohol, 4-amino phenethyl alcohol, o-, p-, m-aminobenzyl alcohol, 2-friend no 4-methyl phenol, 2-friend no 5-methyl phenol, 5-friend no 2-methyl phenol, 2-friend no 4-chlorophenol, 4-friend no 2, 6-dichlorophenol, etc. are mentioned. Moreover, the partial amine ghost of a polyethylene glycol, a polypropylene glycol, or the aliphatic series polyhydric alcohol guided from triol can also be used. When using the partial amine ghost of aliphatic series polyhydric alcohol, number average molecular weight is desirable still more desirable, and 10,000 or less things are 100-2,000. Especially 2-aminoethanol, 3-friend no 1-propanol, 4-friend no 1-butanol, 5-friend no 1-pentanol, 6-friend no 1-hexanol, and 2-(2-aminoethoxy) ethanol \*\* is [ among these ] desirable.

[0021] In manufacture of the hydroxyl-group content polypropylene resin used by this invention, even if the hydroxy group content amine compound enumerated above is independent, or even if it mixes two or more sorts, it can be used.

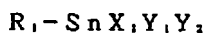
(b) The olefin system elastomer which constitutes the resin constituent used by olefin system elastomer this invention is the copolymer of alpha olefins, such as ethylene, a propylene, butene-1, and a pentene 1, or the copolymer of these and nonconjugated diene. In addition, as nonconjugated diene, a dicyclopentadiene, 1, 4-hexadiene, JISHIKURO octadiene, methylene norbornene, 5-ethylidene 2-norbornene, etc. can be mentioned.

[0022] As an example of an olefin system elastomer, the elastic copolymer of the non-fixed form which uses olefins, such as ethylene propylene copolymer rubber, ethylene butene-1 copolymer rubber, ethylene propylene butene-1 copolymer rubber, an ethylene propylene and nonconjugated diene copolymer rubber, ethylene butene-1 and nonconjugated diene copolymer rubber, and ethylene propylene butene-1, nonconjugated diene copolymer rubber, as a principal component can be mentioned. These can be used, even if independent, or even if it mixes two or more sorts.

[0023] moreover, Mooney viscosity ML 1+4 (100 degrees C) of the above-mentioned olefin system elastomer -- 5-150 -- it is 10-120 preferably. As for the iodine number (whenever [ partial saturation ]) of an olefin system elastomer, it is desirable that it is 16 or less.

(c) The tin compound which constitutes the resin constituent used by the organotin compound and/or tertiary amine compound this invention is the following formula [0024].

[Formula 1]



(式中、R<sub>1</sub>は炭素原子数4～10のアルキル基であり、X<sub>1</sub>は炭素原子数4～10のアルキル基、塩素原子、または水酸基であり、Y<sub>1</sub>及びY<sub>2</sub>は、塩素原子、-OCOR<sub>1</sub>、または水酸基であり、これらは同一であっても異なってもよい。ただし、R<sub>2</sub>はアルキル基、アリール基、アリルアルキル基である。)

[0025] It is the organotin compound come out of and expressed. As such a tin compound, specifically n-C<sub>4</sub>H<sub>9</sub>Sn(OH)<sub>2</sub>Cl, n-C<sub>4</sub>H<sub>9</sub>Sn(OH) Cl<sub>2</sub>, n-C<sub>4</sub>H<sub>9</sub>SnCl<sub>3</sub>, C<sub>8</sub>H<sub>17</sub>Sn(OH)<sub>2</sub>Cl, C<sub>8</sub>H<sub>17</sub>Sn (OH) Cl<sub>2</sub>, C<sub>8</sub>H<sub>17</sub>SnCl<sub>3</sub>, n-C<sub>4</sub>H<sub>9</sub>Sn (OH) 2OCOC<sub>7</sub>H<sub>15</sub>, n-C<sub>4</sub>H<sub>9</sub>Sn (OH) 2OCOC<sub>11</sub>H<sub>23</sub>, n-C<sub>8</sub>H<sub>17</sub>Sn (OH) 2OCOC<sub>7</sub>H<sub>15</sub>, n-C<sub>8</sub>H<sub>17</sub>Sn (OH) 2OCOC<sub>11</sub>H<sub>23</sub>, n-C<sub>4</sub>H<sub>9</sub>Sn 3, (OCOC<sub>7</sub>H<sub>15</sub>) (n-C<sub>4</sub>H<sub>9</sub>) 2Sn (OCOC<sub>11</sub>H<sub>23</sub>)<sub>2</sub>, 2(n-C<sub>8</sub>H<sub>17</sub>) Sn (OCOC<sub>11</sub>H<sub>23</sub>)<sub>2</sub>, 2(n-C<sub>4</sub>H<sub>9</sub>) Sn (OCOCH=CHCOOCH<sub>3</sub>)<sub>2</sub>, and 2(n-C<sub>4</sub>H<sub>9</sub>) Sn(OCOCH=CHCOOCH<sub>2</sub>Ph) 2 grade are used. Among these, n-C<sub>4</sub>H<sub>9</sub>SnCl<sub>3</sub>, 2(n-C<sub>4</sub>H<sub>9</sub>) Sn

(OCOC11H23)2, and 2(n-C8H17) Sn (OCOC11H23)2 are desirable.

[0026] Moreover, as a tertiary amine compound which constitutes the resin constituent used by this invention, dimethyl propylamine, diethyl propylamine, tris (dimethyl aminomethyl) phenol, tetra-guanidine, N, and N-dibutyl ethanolamine and N-MECHIRU N, N-diethanolamine, 1, and 4-diazabicyclo [2.2.2] octane, 1, 8-diazabicyclo [5.4.0]-7-undecene, tetramethyl butanediamine, etc. are mentioned.

[0027] In the resin constituent used by this invention, the tin compound and tertiary amine compound which were enumerated above can be used, whether it mixes two or more sorts even when it is respectively independent, or it mixes a tin compound and a tertiary amine compound.

What is necessary is just to mix the (a) component mentioned above, the (b) component, and the (c) component with various means, in order to manufacture the propylene resin constituent used by manufacture approach this invention of a resin constituent. (a) Especially the sequence of mixing of a component, the (b) component, and the (c) component is not restricted. (a) May mix a component, the (b) component, and the (c) component simultaneously, and (a) The (c) component may be mixed into the mixture of a component and the (b) component, and the (b) component may be mixed into the mixture of the (a) component and the (c) component, and the (a) component may be mixed into the mixture of the (b) component and the (c) component.

[0028] There are various approaches as the manufacture approach of the propylene resin constituent used by this invention. For example, the (c) component is mixed to homogeneity with a tumbler or equipment like a Henschel mixer at the (a) component, the (b) component, and a list, and in case it is processed to a film or a Plastic solid by the extruder and the injection-molding in a plane, it can manufacture. Or a Banbury mixer, a kneader, an extruder, etc. can be used and the approach (the melting kneading method) of mixing the (c) component in the (a) component and (b) component list at the temperature of 300 degrees C or less more than the melting point of raw material resin can also be adopted. Moreover, after mixing the (a) component and the (b) component, the (c) component can also be added and obtained in a fabricating-operation process.

[0029] Among these manufacture approaches, since actuation can pelletize the melting kneading method an easy top in a short time, it is convenient. Although it is desirable to mix each component to homogeneity with a tumbler or equipment like a Henschel mixer beforehand on the occasion of kneading, when required, mixing can be excluded and the approach of carrying out constant feeding to kneading equipment separately, respectively can also be used. Since using the extruder of a multiple spindle as kneading equipment can advance a reaction efficiently, it is desirable.

[0030] It is henceforth. moreover, the thing made for unsaturated carboxylic acid or its anhydride to react to polypropylene under existence of a radical generating agent in the preceding paragraph of an extruder when manufacturing using an extruder -- acid denaturation polypropylene -- manufacturing -- further -- the middle of an extruder -- By adding a hydroxy group content amine compound and supplying the (b) component and the (c) component further, it is also possible to manufacture a propylene resin constituent according to 1 time of an extrusion process, and this manufacture approach is very useful from a manufacturing cost.

[0031] Moreover, the propylene resin constituent used by this invention can add reinforcement, a bulking agent, a coloring agent (a pigment and color), an ultraviolet ray absorbent, a thermostabilizer, a flame retarder, an antioxidant, an antistatic agent, an antifogger, lubricant, a foaming agent, a plasticizer, etc. in a production process or a subsequent processing process if needed in the range which does not spoil the physical properties of resin. Furthermore, according to the application and object, this propylene resin constituent and compatibility can also blend other high thermoplastics with the propylene resin constituent used by this invention. as this example -- a line -- low density polyethylene (LLDPE) and low density polyethylene (LDPE) -- Super-low density polyethylene (VLDPE), high density polyethylene (HDPE), The polymer which consists of one sort of monomers, such as polybutene, the poly isobutene, and Pori (4-MECHIRU 1-pentene), various kinds of ethylene system copolymers (an ethylene-vinylacetate copolymer (EVA) --) An ethylene-vinylalcohol copolymer (EVOH), ethylene and a maleic-anhydride copolymer, Ethylene and an unsaturated-carboxylic-acid



copolymer, and an ethylene alkyl (meta) acrylate copolymer, Furthermore, a styrene butadiene (-styrene) block copolymer and its hydrogenation object, a styrene isoprene (-styrene) block copolymer, its hydrogenation object, etc. are mentioned. One kind of these may be used and may be used combining two or more kinds. Moreover, a copolymer here shows a graft copolymer to random, a block, a random block, and a pan. the molecular weight of these thermoplastics -- usually -- number average molecular weight -- 500-500,000 -- it is the range of 1,000-50,000 preferably. Moreover, the reinforcement used for the propylene resin constituent used by this invention which can use the amount of these thermoplastics used in the range of the 0 - 500 weight section to the propylene resin constituent 100 weight section used by this invention, As an example of a bulking agent, a glass fiber, an asbestos fiber, carbon fiber, A silica fiber, a silica alumina fiber, an alumina fiber, a zirconia fiber, Consolidation fiber, such as boron nitride fiber, silicon nitride fiber, and boron fiber, fumed silica, Inorganic bulking agents, such as clay (aluminum silicate), a glass bead, carbon black, quartz powder, talc (magnesium silicate), titanium oxide, an iron oxide, a calcium carbonate, and the diatom earth, are mentioned.

[0032] For fibrous material, the diameter of average fiber is 5-30 micrometers. Fiber length is 30-50 micrometers. A thing can be used. Since especially a glass fiber raises an interface adhesive property with a propylene resin constituent, and dispersibility, what carried out surface treatment by various coupling agents can be used for it. As a coupling agent, coupling agents, such as a silane system and a titanium system, etc. are usually included. Although an inorganic bulking agent may be used with no processing, what processed the front face with various silane coupling agents, a higher fatty acid, higher-fatty-acid ester, a higher-fatty-acid amide, higher-fatty-acid salts, or other surfactants in order to raise an interface adhesive property with a propylene resin constituent and to raise dispersibility can be used. The mean particle diameter of a desirable inorganic bulking agent is 5.0 micrometers or less, and mean particle diameter is 5.0 micrometers or less still more preferably, and an aspect ratio is five or more. Especially a desirable inorganic bulking agent is talc especially.

[0033] The amount of reinforcement and the bulking agent used is 0 - 40 weight section to the propylene resin constituent 100 weight section. Since shock resistance falls remarkably when reinforcement and/or a bulking agent are included exceeding 40 weight sections, it is not desirable. Furthermore, the desirable range is 5 - 30 weight section. Even if these reinforcement and bulking agents are independent, and they mix two or more sorts, they can be used. 80 - 20% of glass fibers of the inorganic bulking agent in the case of using together especially an inorganic bulking agent and a glass fiber and the ratio of a glass fiber is desirable to 20 - 80 % of the weight of inorganic bulking agents.

(2) Explain the shaping approach for acquiring the shaping approach, next the Plastic solid of this invention. The Plastic solid of this invention can obtain the polar group in a resin constituent (hydroxyl group) by making a Plastic solid front face carry out orientation by making the affinity of the mold front face for shaping, and a polar group into driving force.

[0034] The contact angle of the waterdrop to a Plastic solid front face can be made into the relative scale of the degree of the surface orientation of a polar group, a front face serves as a hydrophilic property with progress of the surface orientation of a polar group, and a waterdrop contact angle falls. The waterdrop contact angle of the propylene resin constituent Plastic solid excellent in the paintwork-ed of this invention is 80 degrees or less, and is 75 degrees or less preferably. Although the surface orientation of a polar group is advancing and it excels in the paintwork-ed amelioration effectiveness so that the contact angle over a front face is small, it is difficult to make a contact angle into less than 60 degrees substantially in the resin constituent used by this invention.

[0035] Although especially definition will not be carried out if it is the shaping approach that surface orientation of a polar group can be effectively advanced as an approach of acquiring the Plastic solid of this invention, as the example \*\* How to acquire a Plastic solid by supplying the ingredient whose surface tension is more than 35 mN/m (20 degrees C) about a resin constituent to the mold for resin shaping which it has on a mold cavity front face, and carrying out a melting press, \*\* Supply the resin by which heating plasticization was carried out after heating beforehand the resin molding die which has the ingredient whose surface tension is more than 35 mN/m (20 degrees C) on a mold cavity front face using high frequency etc. to an elevated temperature 100 degrees C or more. The shaping approaches



and \*\* thermal conductivity which carry out application-of-pressure cooling are below 0.01 cal/cm-sec and \*\* (20 degrees C). And the approach which surface tension supplies the resin constituent by which heating plasticization was carried out in the mold cavity for resin shaping which has the ingredient which is more than 35 mN/m (20 degrees C) on a mold cavity front face, and carries out application-of-pressure cooling is mentioned.

[0036] The approach shown in \*\* among these approaches is very convenient in order to acquire the Plastic solid of this invention, since the front face which was excellent in paintwork-ed, without hardly spoiling productivity in the simple nature of the fabricating operation of resin and a list can be obtained. Hereafter, the approach of the above-mentioned \*\* is further explained to a detail. \*\* The mold for resin shaping which has the ingredient whose thermal conductivity used by the shown approach is below 0.01 cal/cm-sec and \*\* (20 degrees C) and, whose surface tension is more than 35 mN/m (20 degrees C) on a mold cavity front face can be obtained by covering the cavity wall front face of the mold which consists of metals with the ingredient with which are satisfied of the above-mentioned requirements. Since rapid solidification of melting resin advances when the thermal conductivity of the charge of mold facing to be used is bigger than 0.01 cal/cm-sec and \*\*, paintwork-ed and the adhesive amelioration effectiveness are not fully demonstrated, without the orientation by the side of the front face of the polar group in resin advancing efficiently in a fabricating-operation process. But although the amelioration effectiveness can be acquired by setting whenever [ mold temperature ] as an elevated temperature (temperature of the melting point of the propylene system polymer used as a rule of thumb to -50 degrees C or more) when it is an ingredient with larger thermal conductivity than 0.01 cal/cm-sec and \*\*, increase the solidification time amount of resin, shaping effectiveness is made to fall, and it is not desirable. Moreover, although it is desirable since adiabatic efficiency is larger as thermal conductivity is smaller than 0.01 cal/cm-sec and \*\*, generally they are more than 10-5 cal/cm-sec and \*\*.

[0037] moreover, the charge of mold facing to be used -- the surface tension -- Zisman -- it is required for the surface tension in 20 degrees C to be 35 or more mN/m at the appraisal method of the surface tension by law. When surface tension is less than 35 mN/m, it becomes difficult to make coincidence carry out orientation of the polar group to shaping on a Plastic solid front face. Moreover, although surface tension is so desirable that it is large, when balance with the heat conductivity is taken into consideration, generally they are 60 or less mN/m.

[0038] The resin whose surface tension in 20 degrees C is 35 or more mN/m as a suitable thing which fulfills the above-mentioned conditions as a charge of mold facing used in order to acquire the Plastic solid of this invention, a ceramic, resin / ceramic composite material, resin/metal matrix composite, resin / metallic-oxide composite material, ceramic composite material, etc. are mentioned. Moreover, after covering the charge of mold facing, physical or chemical preparation can be performed secondarily, and the surface tension of the charge of mold facing can also be controlled and used.

[0039] Among the above-mentioned charges of mold facing, resin and resin system composite material can obtain the thermal conductivity of ten to 4 (cal/cm-secand\*\*) order, and especially since adiabatic efficiency is large, they are desirable. Since the thermal conductivity of a ceramic system ingredient is 103 order a little more greatly than a resin system ingredient, in order to make the same adiabatic efficiency as a resin ingredient discover, it must make an enveloping layer a little thicker.

[0040] Furthermore, as for the charge of mold facing, it is desirable to give properties, like to excel in the thermal resistance other than the requirements indicated above, that it is strong in a cold energy cycle, to excel in abrasion resistance, that the coat to the body of metal mold can be performed good, that adhesion with the body of metal mold is good, and surface polish can be performed. moreover, about the coat thickness of the charge of mold facing, it is substantially shown in the metal mold outermost layer, and is a thin layer -- buildup of a cooldown delay can be suppressed rattlingly and it is desirable. process conditions, such as thermal conductivity of the ingredient which the thickness of covering of the desirable charge of mold facing for making the surface treatment effectiveness fully discover and not spoiling shaping effectiveness further uses, resin temperature at the time of shaping, a die temperature, and an injection speed of resin, -- since it changes, it is good to decide the thickness of an insulator in consideration of these conditions, molding cycles, etc.

[0041] As an example desirable as an ingredient which covers a metal mold cavity front face, composite material, such as polyester resin, such as polyethylene terephthalate (45 mN/m), an epoxy resin (38 - 45 mN/m), polyimide resin (38 - 42 mN/m) and these resin, a ceramic and a metal, and a metallic oxide, is mentioned. Among these, polyimide resin or especially polyimide resin system composite material is desirable. The surface tension in 20 degrees C of polyimide resin is and is suitable for the range of about 38 to 42 mN/m, when a fluorine atom etc. is not contained.

[0042] Moreover, polyimide resin is convenient especially when covering the metal mold front face of a complicated configuration used with injection molding etc. As for the surface coating ingredient of the mold used in order to acquire the Plastic solid of this invention, it is desirable to have stuck to the metal mold front face firmly repeatedly, so that it can be equal to many shaping. It is desirable to carry out the coat of the metal mold front face by the coat with the sufficient endurance which sticks metal mold enough for that purpose. A complicated metal mold front face is covered with polyimide resin, and the polyamide acid which is the precursor of polyimide resin making it stick firmly can be melted to solvents, such as N-methyl pyrrolidone, and it can apply to a metal mold wall surface, and can carry out most preferably by the approach of heating subsequently and making polyimide resin forming. The polymer of a polyimide precursor has good adhesion with metal mold because of polar groups, such as a carboxyl group, and the thin layer of the polyimide resin stuck to the metal mold front face is obtained by carrying out shaping of the polyimide resin on a metal mold front face.

[0043] Moreover, heating and cooling of a temperature gradient which amount to for every one shaping at the process which supplies the resin constituent by which heating plasticization was carried out in a mold cavity, carries out application-of-pressure cooling, and acquires a Plastic solid, and amount to 100 degrees C or more on a mold front face are repeated. Generally, since the coefficients of thermal expansion of the charge of mold facing and a metal differ greatly, intense stress will occur in the interface of a metal and the charge of mold facing for every shaping. Although it is required that whenever [ breaking strength and breaking extension / both ] should be large, and the adhesion force with a metal should be large as a charge of mold facing which can bear this stress tens of thousands of times, polyimide resin fills these demands and is convenient. The amount polyimide resin of macromolecules of the tough straight chain mold which does not contain the matter which checks adhesion with metals, such as a fluorine, especially is the most desirable.

[0044] Moreover, in order to obtain thermal resistance, the higher one of the glass transition temperature (Tg) of the polyimide resin to be used is desirable. Although Tg of straight chain mold polyimide resin changes with constituents, Tg is desirable still more desirable and 200 degrees C or more are 230 degrees C or more. Although the thermal conductivity of polyimide resin is so desirable that it is small, thermal conductivity is especially desirable and the polyimide resin below 0.002 cal/cm-sec and \*\* can use it.

[0045] The thickness of a polyimide resin layer is moderately chosen in 0.01-2mm. By the thickness of less than 0.01mm, if there is little effectiveness of mold-goods surface amelioration and it exceeds 2mm, the cooling effect of metal mold will fall and shaping effectiveness will fall. It is necessary to thicken thickness of a polyimide resin layer, and is moderately chosen in the range which is 0.01-2mm, so that a die temperature is low. Moreover, it depends for the thickness of a desirable polyimide resin layer also on the fabricating method. For example, in injection molding, the thickness of 0.01-0.5mm is desirable. Furthermore, it is 0.03-0.2mm in thickness preferably. On the other hand, it extrudes and the thickness of 0.1-1mm is desirable in blow molding. In addition, if "the thickness of polyimide" here is placed when a bed depth is not uniform, it means the maximum thickness.

[0046] Furthermore, the large thing of the reinforcement and the ductility of the covering material on the front face of a mold and the amount polyimide resin of straight chain mold macromolecules used is desirable, it is convenient in a cold energy-proof cycle that especially whenever [ breaking extension ] is large, 10% or more is desirable still more desirable, and whenever [ breaking extension ] is 20% or more. Measurement of whenever [ breaking extension ] is performed according to ASTD638. as the suitable example of the amount polyimide resin of macromolecules as a charge of mold facing -- Kapton (a brand name --) the Toray Industries, Inc. make, Tg=428 degree C, and NOBAKKUSU (a brand name

and the Mitsubishi Kasei Corp. make --) Tg=399 degree C and YUPI REXX R (a brand name and the Ube Industries, Ltd. make --) Tg=303, YUPI REXX S (a brand name, the Ube Industries, Ltd. make, Tg=359 degree C) Larc TPI (a brand name, the Mitsui Toatsu Chemicals, Inc. make, Tg=256 degree C), PI2088 (the product made from The Upjohn Co., Tg=342 degree C), and PAI (the product made from Amoco Corp., Tg=230 degree C) are mentioned.

[0047] Moreover, the thermal conductivity indicated above as a mold used in order to acquire the Plastic solid of this invention is below 0.01 cal/cm-sec and \*\* (20 degrees C). and only not only in the metal mold by which the cavity wall front face was covered with the ingredient whose surface tension is more than 35 mN/m (20 degrees C) The whole mold may consist of ingredients whose thermal conductivity is below 0.01 cal/cm-sec and \*\* (20 degrees C) and whose surface tension is more than 35 mN/m (20 degrees C). As this suitable example, an epoxy resin / aluminum composite material (thermal conductivity 0.0034cal/cm-secand\*\* (20 degrees C), trademark Queen NETO NEZ, ZEON Rise make) can be mentioned.

[0048]

[Example] An example and the example of a comparison explain this invention still more concretely below. The manufacture approach of the propylene resin constituent shown in each example, a process condition, and a test method are as being shown below.

The acid denaturation polypropylene which carried out the graft of the maleic anhydride was manufactured by blending organic peroxide with polypropylene, a maleic anhydride, and a list with the loadings shown in the (I) column of the manufacture table 1 of a hydroxyl-group content polypropylene resin (OH-PP), and carrying out melting kneading using a biaxial extruder. After the amount of maleic-anhydride addition in acid denaturation polypropylene performed reprecipitation purification processing by the xylene / acetone system, removed the unreacted maleic-anhydride component and performed the vacuum drying further, it dissolved this purification object in the heat xylene, and asked for it by titrating with a sodium methylate standard solution. The hydroxyl-group content polypropylene resin (OH-PP1 - OH-PP5) was obtained by blending to the acid denaturation polypropylene furthermore obtained with the loadings which show a hydroxy group content amine compound in a table 1 (II), and carrying out melting kneading with a biaxial extruder. Moreover, OH-PP6 was obtained, when a hydroxyl-group content carried out 95 weight sections combination of the polypropylene ethylene block copolymer (PP-4, Asahi Chemical polypropylene M8840) at the hydroxyl-group content polypropylene wax (you MEKKUSU H1210, Made in formation [ Mitsuhiro ]) 5 weight section of 0.9 meq/g and carried out melting kneading with a biaxial extruder.

The quantum of the amount of hydroxyl groups in the measurement hydroxyl-group content polypropylene resin of the hydroxyl-group content in a hydroxyl-group content polypropylene resin was performed by the acetic anhydride / the pyridine method. A detail is shown below.

(1) Hydroxyl-group content polypropylene resin was dissolved in the heat xylene, and it refined by making it reprecipitate in an acetone. The purification sample performed the vacuum drying at 80 degrees C after filtration for 8 hours or more, and removed the solvent.

(2) 3g of purification samples -- xylene: -- it dissolved in 150ml of heating mixed solvents of pyridine =2:1 (volume ratio), the phenol FUTAREN ethanol solution was used and titrated in the indicator with the 0.05NNa methylate standard solution, and the quantum of the acid content in 1g of purification samples was carried out.

(3) 3g of purification samples -- xylene: -- it dissolves in 150ml of heating mixed solvents of pyridine =2:1 (volume ratio), and 5ml (1wt%) of pyridine solutions of an acetic anhydride was added, and they were made to react for 2 hours Then, 1ml of pure water was added and it was made to react for further 2 hours. The phenol FUTAREN ethanol solution was used and titrated in the indicator with the 0.05NNa methylate standard solution after reaction termination, the blank trial was performed by one side, and the consumption of an acetic anhydride was calculated.

(4) The hydroxyl-group content of a hydroxyl-group content polypropylene resin was computed from the acid equivalent in a purification sample, and the consumption of an acetic anhydride. A hydroxyl-group content is the sodium methylate equivalent (it expresses with meq (CH<sub>3</sub> ONa)/g (PP).) contained

in 1g of hydroxyl-group content polypropylene regins.

The test piece for preparation paint film adhesion assessment of the test piece for paint film adhesion assessment was prepared by the following three technique.

- (1) The injection-molding method 1 Melting resin was injected using the injection molding machine (auto shot 50B, FANUC, LTD. make) in the metal mold which performed chrome plating (thermal conductivity  $0.25 \text{ cal/cm-sand}^{**}$  (20 degrees C)) on conditions with a cylinder laying temperature [ of 230 degrees C ], and a metal mold laying temperature of 60-120 degrees C, and the 100mmx100mmx2mm plate Plastic solid was acquired.
- (2) The injection-molding method 2 An injection molding machine (auto shot 50B, product made from FANUC) is used. On conditions with a cylinder laying temperature [ of 230 degrees C ], and a mold laying temperature of 30-120 degrees C Melting resin was injected in the metal mold which covered the cavity mold front face with the polyimide film (brand-name Kapton, the Toray Industries, Inc. make, thermal conductivity  $2.5 \times 10^{-4} \text{ cal/cm-s}$  and  $^{**}$  (20 degrees C), surface tension 38 mN/m (20 degrees C)) with a thickness of 50 micrometers, and the plate Plastic solid was acquired.
- (3) The melting pressing method 1 Press laying temperature was made into 200 degrees C, PET film (brand-name lumiler, Toray Industries, Inc. make, thermal conductivity  $3.6 \times 10^{-4} \text{ cal/cm-s}$  and  $^{**}$  (20 degrees C), surface tension 45 mN/m (20 degrees C)) rolling of the melting resin was carried out, and the 100mmx100mmx2mm plate Plastic solid was acquired.
- (4) The melting pressing method 2 Press laying temperature was made into 200 degrees C, melting resin was rolled out between Teflon films (NAFURON, the NICHIAS CORP. make, thermal conductivity  $6.0 \times 10^{-4} \text{ cal/cm-s}$  and  $^{**}$  (20 degrees C), surface tension 24 mN/m (20 degrees C)), and the 100mmx100mmx2mm plate Plastic solid was acquired.

After washing a waterdrop contact angle measurement Plastic solid front face by pure ethanol, surface ethanol is wiped off, and it was left for 1 hour and made to dry in 23 degrees C. The contact angle of waterdrop 10 minutes after using a syringe for a Plastic solid front face, putting calmly the drop of distilled water with a diameter of 1.5-2.0mm on it under 23 degrees C and the ambient atmosphere of 50 - 60% of humidity and carrying a drop was measured using the contact angle meter (CA-S150 mold, product made from Consonance Interface Science).

Urethane system coating adhesion test (1) After wiping off the plane surface with a thickness of 2mm which pretreated, could be burned and carried out processing shaping with the cloth to which impregnation of the ethanol was carried out and drying it in 20-25 degrees C for 1 hour or more, it sprayed using the 2 liquid type urethane system coating (RETAN PG 60, Kansai Paint Co., Ltd. make) (30-40 micrometers in thickness). Then, after leaving it for 30 minutes in 20-25 degrees C, it processed by the ability being burned at 80 degrees C by the hot blast circuit system desiccation in a plane for 1 hour. After ending baking processing, after leaving it for further 24 hours, the paint film friction test shown below performed paint film adhesion assessment. (2) After the test piece which carried out paint film friction test paint was immersed in 40-degree C warm water for 240 hours, the cutter knife was used on the paint film side, the parallel lines which go direct were drawn at intervals of every 11mm [ 1 ], and the 100 squares were made. Cellophane adhesive tape (JIS-Z1522) was fully stuck by pressure on it, and it kept at a paint film side and about 30 degrees, and pulled and removed at a stretch to the front, and the condition of the part surrounded by the squares was observed. The result was expressed with a residual number / 100.

[0049]

[Examples 1-3] The propylene resin constituent was obtained by carrying out melting kneading of the hydroxyl-group content polypropylene regin (OH-PP1) 70 weight section, the polyolefine system elastomer (EPR1) 30 weight section, and the organotin compound (DBTDL) 0.1 weight section with a biaxial extruder. It fabricated by the process condition which shows these resin constituents in a table 2, and the waterdrop surface contact angle of a Plastic solid and the adhesion of an urethane system coating which were acquired were evaluated. A result is shown in the table 2 lower part. The paint film adhesion in which the contact angle of waterdrop was excellent in the Plastic solid 80 degrees or less was acquired.

[0050]

[The examples 1 and 2 of a comparison] It fabricated by the process condition which shows the propylene resin constituent used in the examples 1-3 in a table 2, and the waterdrop surface contact angle of a Plastic solid and the adhesion of an urethane system coating which were acquired were evaluated. A result is shown in the table 2 lower part. In the Plastic solid 80 degrees or more, the paint film exfoliated [ the contact angle of waterdrop ] easily.

[0051]

[Examples 4-10] It blended with the loadings which show a hydroxyl-group content polypropylene resin (a), a polyolefine system elastomer (b), an organotin compound and/or a tertiary amine compound (c), other thermoplastics, and talc in a table 3, and the propylene resin constituent was obtained by carrying out melting kneading with a biaxial extruder. It fabricated by the approach of showing these resin constituents in the injection-molding method 2, and the plate Plastic solid was acquired. The waterdrop surface contact angle of a Plastic solid and the adhesion of an urethane system coating which were acquired were evaluated. The paint film adhesion of a Plastic solid was extremely excellent.

[0052]

[The examples 3-5 of a comparison] The test piece for paint film assessment was fabricated by the approach shown in the injection-molding method 1 from the propylene resin constituent used in the examples 4, 5, and 6, and the waterdrop surface contact angle of a Plastic solid and the adhesion of an urethane system coating which were acquired were evaluated. A result is shown in the table 3 lower part.

[0053]

[A table 1]

				OH-PP1	OH-PP2	OH-PP3	OH-PP4	OH-PP5	OH-PP6
水酸基含有ポリプロピレン系樹脂	(I) 酸変性ポリプロピレン	ポリプロピレン	PP-1	100					
			PP-2		100		100	100	
			PP-3			100			
		無水マレイン酸		1.0	1.0	1.0	1.0	1.5	
		パーオキサイド		0.1	0.1	0.1	0.1	0.2	
		無水マレイン酸付加率(wt%)		0.45	0.56	0.60	0.56	1.21	
	(II) アミノアルコール	アミノアルコール		1.0	1.25	1.25		1.5	
		アミノアルコール					1.2		
	ポリプロピレン		PP-4						85
	水酸基含有PPワックス								5
	水酸基含有量 (10 <sup>-2</sup> meq/g )			2.20	2.50	2.80	2.10	4.21	1.70

(注) 表中で単位が指定されていない数値に対する単位はすべて重量部である。

[0054]

[A table 2]

		実施例1	実施例2	実施例3	比較例1	比較例2
成形条件	成形方法	溶融プレス法1	射出成形法1	射出成形法2	射出成形法1	溶融プレス法2
	成形基板材料	PET	Crメッキ	ポリイミド	Crメッキ	テフロン
	基板温度 (°C)	200	110	40	60	200
水滴表面接触角 (° (23°C))		65	75	72	82	90
塗膜密着性 (残存数/100)		100/100	100/100	100/100	10/100	0/100

用いた樹脂組成物の組成: OH-PP1/EPR1/DBTDL=70/30/0.1 (重量部)

[0055]

[A table 3]

		実施例4	実施例5	実施例6	実施例7	実施例8	実施例9	実施例10	比較例3	比較例4	比較例5
プロピレン系樹脂組成物	(a) 水素含有ポリプロピレン系樹脂	OH-PP2	90	70		70				90	70
		OH-PP3			60						60
		OH-PP4				50					
		OH-PP5					30				
		OH-PP6						80			
	(b) オレフィン系エラストマー	EPR-1	10	30		30	50	70	20	10	30
		EPR-2			40						40
	(c) 有機スズ及び/または第3級アミン化合物	DBTDL	0.1	0.1		0.1	0.2	0.2	0.2	0.1	0.1
		DABCO			0.1	0.1					0.1
	その他の熱可塑性樹脂	HDPE					10				
		LDPE						15			
		SEBS				15					
	強化材	タルク			15		10				
成形条件	成形方法	射出法2	射出法2	射出法2	射出法2	射出法2	射出法2	射出法2	射出法1	射出法1	射出法1
	基板温度 (°C)	60	40	80	60	30	60	90	60	80	40
水滴表面接触角 (° (23°C))		72	70	66	72	72	68	74	82	83	84
塗膜密着性 (残存数/100)		100/100	100/100	100/100	100/100	100/100	100/100	100/100	0/100	10/100	20/100

(注) 表中で単位が指定されていない数値に対する単位はすべて重量部である。

[0056] The code in a table 1 - a table 3 is as follows.

Polypropylene O gay polypropylene (PP-1)

Asahi Chemical polypropylene E1100MFR:0.5g /, 10min (ASTM D1238)

O Polypropylene ethylene block copolymer (PP-2)

Asahi Chemical polypropylene M7100MFR:0.5g /, 10min (ASTM D1238)

O Polypropylene ethylene random copolymer (PP-3)

Asahi Chemical polypropylene E3100MFR:0.5g /, 10min (ASTM D1238)

O Polypropylene ethylene block copolymer (PP-4)

Asahi Chemical polypropylene M8840MFR:40g /, 10min (ASTM D1238)

Organic peroxide (peroxide) O2, the 5-dimethyl 2, 5-G (tert-butyl peroxide) hexane maleic-anhydride O

Nippon Oil & Fats Crystal man hydroxy group content amine compound O 2-aminoethanol (amino alcohol 1)

O 2-(2-aminoethoxy) ethanol (amino alcohol 2)

hydroxyl-group content polypropylene wax O Mitsuhiro -- formation You MEKKUSU H1210 number-average-molecular-weight 4,000 and hydroxyl value:50 mgKOH/g olefin system elastomer O ethylene

propylene copolymer rubber (EPR-1)

Exon chemistry VISTALON805 Mooney viscosity ML 1+4 (100 degrees C):53, ethylene content:77wt%O ethylene propylene copolymer rubber (EPR-2)

Exon chemistry VISTALON878 Mooney viscosity ML 1+4 (100 degrees C):76, ethylene content:54wt% organotin compound O dibutyltin dilaurate (DBTDL)

Tertiary amine compound O diazabicyclo [ 1 and 4- ] [2.2.2] octane (DABCO)

Other thermoplastics O high density polyethylene (HDPE)

Asahi Chemical SANYO ENGINEERING & CONSTRUCTION-HD J340MFR:7g /, 10min (ASTM D1238)

O Low density polyethylene (LDPE)

Asahi Chemical SANYO ENGINEERING & CONSTRUCTION-LD M6520MFR:20g /, 10min (ASTM D1238)

O The hydrogenation object of styrene Butadiene Styrene (SEBS)

Asahi Chemical tough tech H1052MFR:12g /, 10min (ASTM D1238)

Styrene content: 19wt% reinforcement (talc) O Japan talc Micro ace p4 [0057]

[Effect of the Invention] The propylene resin Plastic solid of this invention is excellent in the paintwork-  
ed over an urethane system coating. An urethane system coating is a coating which generally forms a  
paint film by the reaction of the poly isocyanate and a polyol compound, has a 1 liquid type and a 2  
liquid type, and also has the powder coatings using block-type isocyanate. The propylene resin form  
acquired by this invention is excellent in the adhesion also to which [ these ] urethane system coating.  
Since the Plastic solid furthermore acquired by this invention has the outstanding surface smooth nature,  
especially the feeling of a result after paint is excellent. Moreover, since the mold for shaping used in  
order to acquire the Plastic solid of this invention is conventionally applicable to the well-known  
fabricating-operation method of arbitration, for example, injection molding, compression molding,  
extrusion molding (sheet forming, blow molding), etc., the Plastic solid of various configurations can be  
acquired easily and the practically useful product covered very variously can be made.

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[Translation done.]



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1. This document has been translated by computer. So the translation may not reflect the original precisely.
2. \*\*\*\* shows the word which can not be translated.
3. In the drawings, any words are not translated.

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CLAIMS

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[Claim(s)]

[Claim 1] The propylene resin Plastic solid characterized by being the Plastic solid of the resin constituent which consists of the following (a), (b), and (c) components, and the contact angle of the waterdrop to this Plastic solid front face being 80 degrees or less.

(a) component: -- hydroxyl-group content polypropylene resin [ in which a hydroxyl group is contained below 50 meq/g (PP) more than  $0.1 \times 10^{-2}$  meq/g (PP) ] 99.9 - 10 weight (sections b) component: olefin system elastomer 0.1 - 90 weight section (c) component: -- an organotin compound and/or tertiary amine compound a total of 100 weight sections of the (a) component and the (b) component -- receiving -- 0.01 - 10 weight section [claim 2] The manufacture approach of the propylene resin Plastic solid characterized by carrying out heating plasticization, supplying the resin constituent which consists of the following (a), (b), and (c) components, and carrying out application-of-pressure cooling into the mold cavity for resin shaping which has the ingredient whose thermal conductivity is below 0.01 cal/cm-sec and \*\* (20 degrees C) and, whose surface tension is more than 35 mN/m (20 degrees C) on a mold cavity front face.

(a) component: -- hydroxyl-group content polypropylene resin [ in which a hydroxyl group is contained below 50 meq/g (PP) more than  $0.1 \times 10^{-2}$  meq/g (PP) ] 99.9 - 10 weight (sections b) component: olefin system elastomer 0.1 - 90 weight section (c) component: -- an organotin compound and/or tertiary amine compound a total of 100 weight sections of the (a) component and the (b) component -- receiving -- 0.01 - 10 weight section

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[Translation done.]